Inner demagnetizing factor in polymer bonded soft magnetic composites

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1. Introduction

Plastics filled with soft magnetic particles (polymer bonded soft magnetic composites - PBSMC) for low frequency applications are attracting more and more the interest of industry. Objectives of these inventions are the development of soft magnetic composites for low frequency applications with high relative permeabilities, low core losses and low coercivities, capable of processing with typical plastics processing procedures like extrusion and injection molding. In past various works have been done to characterize magnetic composites. Theoretical and experimental works show an increase of permeability and decrease of coercivity of the PBSMC with increasing filler fraction of soft magnetic particles.

However, to characterize magnetic properties as permeability, remanence or the actual applied magnetic field, knowledge of the demagnetizing factor is unavoidable. For a bulk material approaches, tables or exact calculations are existing but for materials consisting of a non-magnetic matrix and a magnetic filler, only few number of published works can be found [1-6].

2. Theoretical

Except for a closed toroidal core of homogeneous full material at every magnetic core an inner demagnetizing field arises, which reduces the applied field H_0 to $H_{int} = H_0 - N \cdot M$ (the inner magnetic field in the specimen is H_{int}, the demagnetizing factor is N and the magnetization is M).

The demagnetizing factor N_{total} of a PBSMC depends on the inner demagnetizing factor N_i (depending on particle content, shape and size distribution) and on the external, geometric demagnetizing factor N_{geo} (depending on the shape of the specimen). The total demagnetizing factor is the sum of geometrical and inner demagnetizing factor $N_{total} = N_{geo} + N_i$ [7,8]. The external demagnetization factor

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of rotationally symmetrical ellipsoids submited to an uniform external field is exactly calculable since in this sample form a homogeneous magnetization occurs [8].

The external demagnetizing factor can be determined by comparison of a measured hysteresis curve with an ideal hysteresis curve. The slope of an ideal hysteresis curve for small magnetization M is infinite and therefore the inner demagnetizing field is zero. By estimating the angle α between an experimental and an ideal hysteresis curve at low magnetization M, N_{total} can be calculated by [7]:

$$\tan(\alpha) = \frac{H}{M} = N_{total} \tag{1}$$

It is an experimental fact that inner demagnetizing effects turn to outer effects when the fraction in magnetic matter goes through a particular value [5]. For heterogeneous and randomly structured materials, like PBSMCs the external demagnetization factor rises steeply in a linear way from 0 to N_{geo} between filler fractions from 20 to 40-vol.% due to arising cooperative phenomena in the composite [6].

Furthermore for an heterogeneous material an inner demagnetizing factor occurs which is not dependent on sample's geometry. Since a PBSMC consists of soft magnetic particles that are randomly dispersed in a polymer matrix, this body does not consist of 100% homogeneous soft magnetic material and the polymer matrix represents gaps in the magnetic material. Around the single particles in a magnetic particulate filled polymer therefore additional demagnetizing fields occur. Thus, inner demagnetizing fields also arise in toroidal cores.

For a composite material with a filler fraction of $C \to 0$ the demagnetizing factor of the compound equals the demagnetizing factor N_P of a single particle. The calculation of demagnetizing factors of simple geometrical shapes (spheres, cylinders, etc.) is given in Kneller [8]. The demagnetizing factor of a sphere is $N_{sphere} = 1/3$ and that of a cylinder with a length/diameter ratio of 1 is approximately $N_{cyl} \approx 0.27$ at its center [9].

The fact that at low filler fractions the particles are magnetically independent each other and that the total demagnetization factor for every single particle must be considered (for spherical particles $N_P = 1/3$) is responsible for that behavior. In the case of magnetic independency of particles only this internal demagnetization factor occurs. Cooperative phenomena in the composite occur and reduce the demagnetization field through beginning interactions between the particles [6]. With increasing filler fraction the field strength of the inner demagnetizing field is decreased and should converge to $N_i \approx 0$ for a filler fraction of $C \to 1$.

Mixing laws are often used to get the effective properties of composite materials as a function of the intrinsic properties of their components. Among them, the Effective Medium Theory (EMT) based on the works of Bruggeman and Landauer [10], show the salient interest to predict the existence of a percolation threshold in the electrical behaviour of heterogeneous media. We have shown that a similar approach leads to a satisfying description of interactions between magnetic inclusions in disordered heterostructures. Our approach rules experimental variations of the inner and outer demagnetizing effects shown by heterostructures. The magnetic load of the composite medium is C, and the magnetic percolation threshold is C_p. When $C < C_p$, magnetic gaps, which originate from the magnetic dilution, may cause a cut-off of the magnetic flux-paths in the composite medium, leading to inner demagnetizing effects only. When $C > C_p$, the delocalization of the magnetic poles leads the outer demagnetizing factor of the sample to switch from zero to its maximum value.

A detailed elaboration of the used EMT can be found in [5, 6]. Starting from a self consistent law, we found that the effective permeability μ_e of randomly ellipsoids of identical shapes - characterized by their permeability μ_i and there tensorial shape factor $\tilde{N}$ with the diagonal components (N_x, N_y, N_z) - immersed in a non-magnetic matrix, and in the field of a dipolar approximation, obeys to

$$\sum_{\alpha=x,y,z} \left(\begin{array}{l} \frac{C}{3} \cdot \left[\frac{(\mu_i - \mu_e)(1-N_\alpha)}{\mu_e + (\mu_i - \mu_e)N_\alpha} \right] + \\ \frac{(1-C)}{3} \cdot \left[\frac{(1-\mu_e)(1-N_\alpha)}{\mu_e + (1-\mu_e)N_\alpha} \right] \end{array} \right) = 0 \tag{2}$$

Under only slightly restrictive conditions (the shape of the inclusion is assumed to be ellipsoidal, and nearly spheroidal, that allows to write $N_x \cong N_y \cong 1 - N_z$), and under the assumption that the magnetic contrast between the components is not too high (numerical simulations show that this assumption is relevant when $\mu_i \leq 15$) eq. (2) writes in the more versatile scalar form [5]:

$$(1-N)\cdot \chi_e^2 + [1 + (N-C)\chi_i]\cdot \chi_e - C\cdot \chi_e = 0 \tag{3}$$

where $N = \frac{2 + 3\delta N_z(1-N_z)}{6 + \delta + 3\delta N_z}$ and $\delta = \mu_i / \mu_e - 1$

with effective susceptibility $_e$ and particle's intrinsic susceptibility $_i$.

3. Experimental

Measurements of the demagnetizing factor where performed with a computer controlled hysteresis measurement system similar to that one described in [11, 12]. For all materials measurements were carried out at the maximum possible magnetic field of approximately $H \approx 50$ *kA/m*. Measurements were done at a magnetizing frequency of $f = 1$ *Hz*. Ring-shaped samples were placed in a HP APC7 coaxial line and the susceptibilities got with a HP

8720 network analyzer.

As magnetic materials an iron-silicon powder (FeSi6.8, Höganäs AB, Sweden) with a mean particle diameter of d_{FeSi} = 136 μm, irregular shaped iron powder (Fe, ASC100.29, Höganäs AB, Sweden) with a mean diameter of d_{Fe} = 88 μm and needle shaped nano-crystalline $Fe_{73.5}Si_{15.5}B_7Cu_1Nb_3$ powder (Finemet, Arcelor S.A., Luxembourg) with a mean diameter of d = 35 μm. Micrographs of FeSi6.8, Fe and Finemet can be seen in fig. 1. As polymeric material a polypropylene (Moplen-EP540N, Basell Polyolefine GmbH, Germany) was chosen. Iron and iron-silicon powders and the preparation methods of the PP-magnetic powder-composites are described in more detail in [13, 14]. Composites from 7 to 85-vol.% filler fraction were prepared.

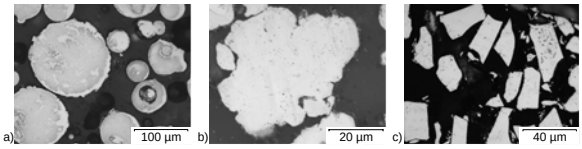


Fig. 1: Micrographs of used soft magnetic powders: a) gas atomized FeSi6.8, b) water atomized Fe, c) milled $Fe_{73.5}Si_{15.5}B_7Cu_1Nb_3$ ribbons

The demagnetizing factors were estimated as described in [3, 7] and referred to the content of magnetic material in the PBSMC by multiplying with filler fraction. The geometrical demagnetizing factor N_{geo} of the PBSMC was first estimated following [9] to be N_{geo} = 0.004 and afterwards experimentally determined.

To minimize inner demagnetizing effects in measurements of geometrical demagnetizing factor in experimental determination of N_{geo} a sample of a FeCo-alloy with same dimensions as the PBSMC samples was used. Thus, geometrical demagnetizing factor was estimated to be N_{geo} = 0.004.

4. Results and discussion

In fig. 2 a comparison of inner demagnetizing factors of own measurement values of polypropylene filled with spherical FeSi particles, with irregular shaped iron particles and with nano-crystalline $Fe_{73.5}Si_{15.5}B_7Cu_1Nb_3$ particles dependent on the particle filler fraction with data from literature can be seen [1-3].

At first it is recognizable that there are three groups of measured data, which can be described by three fit curves.

The first group of measured values can be described by the function: $N_i(C) = 1/3exp(-3C)$. At a filler fraction of C = 0 the curve intersects the ordinate at N_i = 1/3 which is the demagnetization factor of a sphere. As can be seen this function describes satisfactory the filler fraction dependence of composites filled with spherical particles of FeSi and Fe. The iron particles used by Forrer had a diameter of 1 mm, while the FeSi particles had a mean diameter of $d \approx$ 136 μm. But also composites with irregular shaped Fe-particles show the same behavior as composites with spherical particles.

The second group of measurement values can be described by the function $N_i(C) = 0.288exp(-3C)-0.004$. On this function measurement values of permalloy parallelepipeds and cut iron wires found by Forrer [1]. Unfortunately

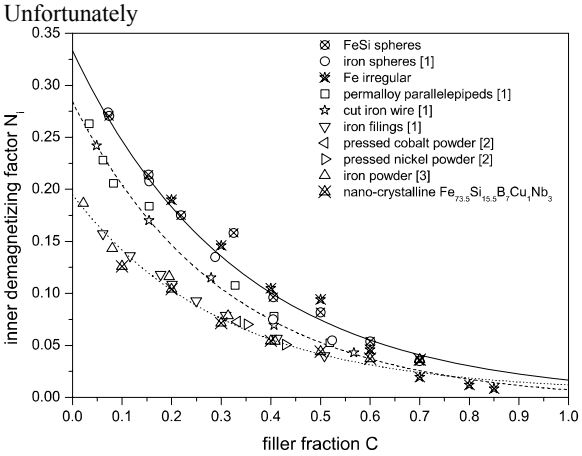


Fig. 2: Comparison of measured inner demagnetizing factors of different composites at various magnetic particle fractions. Symbols marked with a cross are own measurements, other data are from Forrer [1], Gerlach [2] and Kranz [3], fit lines are described in the text.

Forrer did not give any further specifications of his magnetic material. However the inner demagnetizing factor of $N_i \approx 0.28$ at a filler fraction of C = 0 can be found for cuboids of a length to width ratio of 1 : 1.25 [15].

The third group of results can be described by the function $N_i(C) = 0.19exp(-3.3C)+0.005$. This function of measurement values is congruent with values of milled nano-crystalline $Fe_{73.5}Si_{15.5}B_7Cu_1Nb_3$ ribbons, iron files by Forrer [1], iron powder used by Kranz [3] and nickel powder used by Gerlach [2]. It is astonishing that all materials show the same curse of demagnetizing factor versus filler fraction. Since there are no data about the shape of the used powders given, one can not give an explanation for that coincidence. Either all materials have a similar shape or unoriented materials of irregular, longish shape show a total demagnetizing factor of about 0.19. To validate the measurements of the demagnetizing factors a blend of $Fe_{73.5}Si_{15.5}B_7Cu_1Nb_3$ and iron-silicon with the ratio 1:1 has been measured. The resulting demagnetizing factor was exactly the average of both single measured demagnetizing factors.

However, it has to be pointed out that the fit function shown in fig. 2 has no physical meaning. Nevertheless, the fit functions and also measured values at high filler fractions show inner demagnetizing factors larger than N_i = 0. Because the crystallites of a 100% magnetic sample are usually misaligned in respect to the axis of the macroscopic sample always a non-zero inner demagnetizing factor

appears [16].

The fact that the internal demagnetizing factor for $C = 1$ is not zero, in contrast to theoretical considerations, can be explained by referring to [3, 7, 16]: the internal demagnetizing factor not only depends on the filler fraction but also on the shape and distribution of the crystallites in the particles itself as well as of varying internal tensions. For example the internal demagnetizing factor of cold worked materials can be much higher than in a relaxed sample. I.e. also a bulk material may show an inner demagnetizing field. Moreover it seems obvious that, for filling fractions above $C = 0.6$, the shape of the inclusions (particle or clusters) is modified by the manufacturing process.

With the measured properties of the materials one always measures the properties of particles. A sample material of 100% soft magnetic filler would still be 100% particles and show boundaries of the particles, grain boundaries and possible internal tension evoked by quenching or milling.

During the powder mixing process**, it was not attempted to avoid aggregation of particles**. It has been shown [17] that when the particle concentration in a given mixture is increasing, aggregates are rapidly formed. Their shapes are continuously changing from chains to spheres up to the concentration unity where the medium becomes isotropic. Moreover, magnetic interactions, and particularly long range dipolar interactions, are not prevented by an electrical insulating coating of the magnetic particles. Therefore, taking the aggregation process of particles into account, the statistical distribution of aggregates shape with the volume fraction has been established [17]. It is shown in particular, that the shape factor that appears in the used EMT may not be necessarily equal to 1/3 in the very low filling fraction range even for randomly dispersed (non spherical) particles.

Only results got from FeSi spheres where compared with the theoretic consideration and are presented here (fig. 3). Using the variation of the susceptibility shown in fig. 3 we found μ_i close to 4. This value, which is typical for a rotational behaviour, is in accordance with results got from susceptibility measurements obtained for ring-shaped samples of soft materials[6], from which we deduced $\mu_i = 5$ (fig. 3) and $N_z = 0.29$ which is close to the theoretical value of 1/3 for spherical particles. The variation of the demagnetizing coefficient N_z is easily obtainedfrom eq. (3), where μ_e is the permeability of the sample (fig.2) and where μ_i is the intrinsic rotational permeability of the magnetic component.

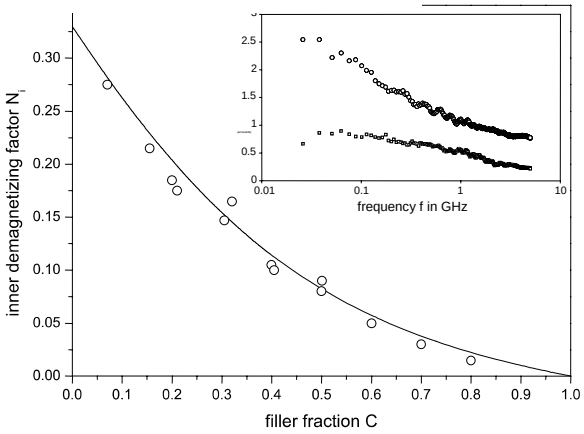
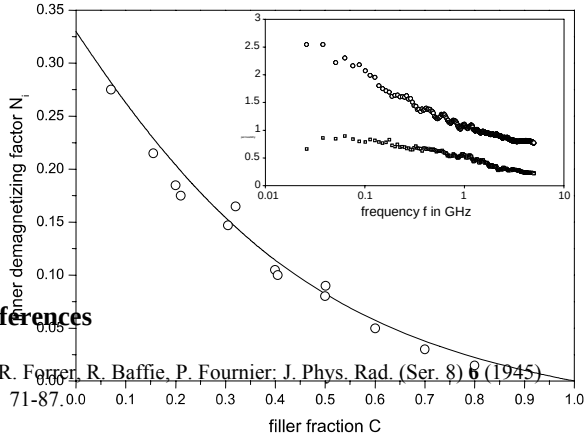


Fig. 3: demagnetizing factors vs. *C*, for the FeSi samples are data extracted from fig. 2 and fitted by eq. (3).



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